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ONR Grant No. N00014-90-J-4077

Relaxor Ferroelectrics for Electrostrictive Transducers

Quarterly Report

Electrostrictive Strain/Dielectric Properties of Relaxor Ferroelectrics

Summer 1991

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Introduction

The electrostrictive actuator and nonresonant transducer capabilities of relaxor ferroelectrics have been of practical interest for the last decade.[1,2] Displacement transducers from materials which have large electrostrictive effects are advantageous for several reasons:

(i) The field-induced deformations (strains) are more stable than in piezoelectric ceramics (relaxation processes are minor and they do not evidence aging from macrodomain-defect interactions).

(ii) Hysteresis in the strain-field dependence is reduced over a wide temperature range. The original dimension is returned to rapidly after removal of the polarizing field (reduced creep and strain offset).

(iii) Thermal expansion effects are quite small (~ 10^{-6}/°K) in the relevant temperature range of maximum dielectric permittivity.

This report is intended to update progress on the most prominent compositions from the selected families of relaxors and present information on their dielectric and electromechanical properties of interest. A wide temperature range has been investigated for dielectric permittivity, K(T), strain-E field and polarization-E field response. Strain levels achieved for a constant E field level and also the E field needed to reach a constant maximum strain level were also recorded and tabulated.

Results for PMN-PT (with modifications) and the PLZT (9,10,11/65/35) compositions are emphasized in this quarterly report and a summary of their electrostrictive properties are included. A future work section will discuss additional experimental undertakings and other compositional systems of relaxor ferroelectrics to be investigated

Experimental Procedure

Electromechanical strains and dielectric polarizations were concurrently measured for cycling at modest frequencies (<1 Hz) around their high-field electric hysteresis loops. As described in the previous quarterly report[3] dielectric constants ,K(T), were measured by a capacitance bridge and a strain gauge method was used for the determination of the electromechanical properties of high-field electrostriction for PMN, .98 PMN-.02 PT, and .95 PMN-.05 PT:1% La, and PLZT (9,10,11/65/35) ceramic samples. The LVDT (linear voltage differential transformer) method was also used to confirm the strain behavior.

Results and Discussion

Relaxor ferroelectrics in the PMN-PT based system can be put into three groups according to their dielectric/polarization behavior: Region I - Electrostrictive, where the S-E and P-E hysteresis is virtually negligible and the strain is proportional to the polarization squared and with reduced ageing or creep effects; Region II - Micro-Macro Domain Region, where large field-induced strains are possible and the strain hysteresis is still not large and little change in the creep/setpoint for strain is observed; Region III - Macrodomain, in the temperature range where remanent polarization exists and concommitant remanent strain and significant dielectric and strain hysteresis occurs.

The compositions PMN, .98PMN- .02PT, and .95PMN- .05PT : 1% La all exhibit large frequency dependent dielectric constant peaks as shown in Figs. 1 - 3 and with Tm at just below or just above 0° C. With only moderate dielectric loss peaks (tan $\delta<0.10$) and large values of dielectric constant, the sensitivity of the strain -E field response should be reasonably high in the (-10° to +40° C) temperature range.

The transverse strain (negative) measured as a function of temperature are shown in Fig. 4 for these compositions at low frequency (0.1 Hz) and field levels of 10 and 20 Kv/cm. Table I lists strain levels for the same compositions and field levels of 10 and 20 Kv/cm. Figs. 5, 6, and 7 indicate the electric field strain hysteresis response for these compositions as a function of temperature.

The strain response of the various electrostrictive materials followed expected behavior[4,5]. As reported in Table I and shown in Fig. 4, the transverse electrostrictive strain increased with decreasing temperature, and increasing dielectric constant maximum was approached. Since the measurement frequency was 0.1 Hz, the effective T_m was lower than that shown in Figs. 1- 3, and hence electrostrictive behavior was observed well below these reported T_m values. As shown, minimal hysteresis is observed until well below T_m, whereupon micro-macrodomain switching occurs. It should be noted that the minimal hysteresis shown, particularly for PMN-PT:La, is not indicative of the sample but of the electrode-strain gauge interface. Hysteresis in Region I (electrostrictive) may also be the result of dielectric-electrode effects such as space charge and overall non E-field/polarization homogeneity. This observation is substantiated by the fact that the same level of hysteresis occurs over a wide temperature range well above T_m.

The relaxor PLZT compositions were examined over a similar temperature range via the same methods and strain levels in accord with previous researchers' result[6] were obtained. Figs. 8, 9, and 10 indicate the level of transverse strain achieved at the field levels of 10 KV/cm and 20 KV/cm. The strain for PLZT 9/65/35 is considerably higher than for either PLZT 10/65/35 or

11/65/35. The field induced strain levels for PLZT 10/65/35 and 11/65/35 are nearly the same at higher field levels.

The measured T_m values of 84.4° C, 53.2° C, and 39.0° C for PLZT (9,10,11/65/35), respectively, indicates that in the higher end of the measured temperature range these materials are all in the micro-macrodomain region II and at lower temperatures (especially for 9/65/35) the response becomes more like a macrodomain polar ferroelectric. This leads to the "butterfly" piezoelectric ceramic-like strain hysteresis loops seen in PLZT below T_d.

In order to assess the utility of the PLZT materials' important device parameters, such as the amount of electric field needed to induce a constant maximum required strain level or the amount of strain induced at a constant electric field should be determined. Fig. 11 shows the electric field strain level (3×10^{-4}) at various temperatures. Figure 12 shows the strain level (6×10^{-4}) and required electric fields at various temperatures for PLZT (9/65/35). Tabulated values for these compositions are shown in Tables 3 and 4.

Fig. 13 shows the transverse strain with a constant maximum electric field (27.5 Kv/cm) for PLZT (10/65/35). Increasing strain levels and hysteresis are observed with decreasing temperature

Summary and Future Work

As expected the electrostrictive strain behavior of PMN-based materials is dependent on the temperature in relation to T_m. By simply fabricating relaxor ferroelectric materials with Tm near 0° C, electrostrictive behavior with minimal hysteresis was observed. Only well below Tm approaching T_d does hysteresis play a significant role due to micro-macro domain switching. Though shifting T_m downward effectively offers electrostrictive behavior at lower temperatures, the intrinsic decrease in Kmax leads to slightly less strain.

The strain response as a function of electric field in PLZT compositions is quite large and exceeds the (.03%) level set as a minimum standard for the temperature range. The amount of strain and the strain offset and hysteresis is quite sensitive to the amount of La added.

A further development of the electromechanical properties of the PMN-PT system and Pb(Mg, Ta)O3- PT compositions and its modification with Sr and charge compensating aliovalent ions K and La remains for completion, of the three initial proposed families of electrostrictive relaxors. Analysis of the more promising PLZT, PMN-PT/(La), and PNN-PZT compositions optimized for structure-composition leading to large electrostrictive response and good mechanical driving strength. Prestress testing of the mechanical driving force for these samples under loading will be undertaken.

The $Ba(Ti, Sn)O_3$ [7] family of electrostrictors are also of interest because of the following possible advantages:

-large diffuse dielectric constant

-larger strain at lower E-field level, due to:

-nearly linear S-E response (possibly due to nonlinear dielectric response)

and will be investigated.

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Table 2. Strain response for relaxor PLZT compositions of various temperatures and electric fields [PLZT (9,10,11/65/35)].

PLZT Composition	Temp.	Transverse Strain ($x10^{-4}$) 10 kV/cm	20 kV/cm	30 kV/cm	Hysteresis
(11/65/35)	24°	1.9	3.15		Minimal
	18°	2.25	3.20		"
	13°	2.2	3.50		"
	10°	2.3	3.6		Modest
	0°	2.55	3.75		"
	-7°	2.70	3.95		"
(10/65/35)	23°	.85	3.15	4.35	Minimal
	4°	1.1	3.9	5.2	"
	-16°	1.3	4.6	6.0	Significant
(9/65/35)	22°	5.8	7.5		Modest
	17°	5.8			"
	14°	5.9			"
	12°	6.1			Significant
	10°	6.15			"
	6°	6.30			"
	4°	6.55			"
	1°	6.70			"
	-36°	9.40			"

Table 3.

PLZT 11/65/35

Temperature	E-field to achieve 300 με [kV/cm]
+21°C	15.4
0°C	13.8
-11°C	11.2
-21°C	9.4

Table 4

PLZT 9/65/35

Temperature	E-field to achieve 600 με [kV/cm]
27	12.5
23	10.6
14	10.1
8	9.8
-3	9.5
-10	8.8

References

[1] S.J. Jang, Ph.D Thesis, The Pennsylvania State University (1979).

[2] K. Uchino, S. Nomura, L.E. Cross, S.J. Jang, and R.E. Newnham, "Electrostrictive Effect in Lead Magnesium Niobate Single Crystals," J. Appl. Phys., Vol. 51, No. 2, pp. 1142-45 (1980).

[3] Report on ONR Grant No. N00014-90-J-4077, March/April 1991.

[4] M. Ealy, C. Wheeler, " A Standardized Electrodisplacive Transducer," SPIE Symp. on Aerospace Sensing, Orlando, FL March (1989).

[5] T.R Shrout and J. Fielding, Jr., "Relaxor Ferroelectric Materials," Proc. IEEE Ultrasonics Symp., Dec. (1990).

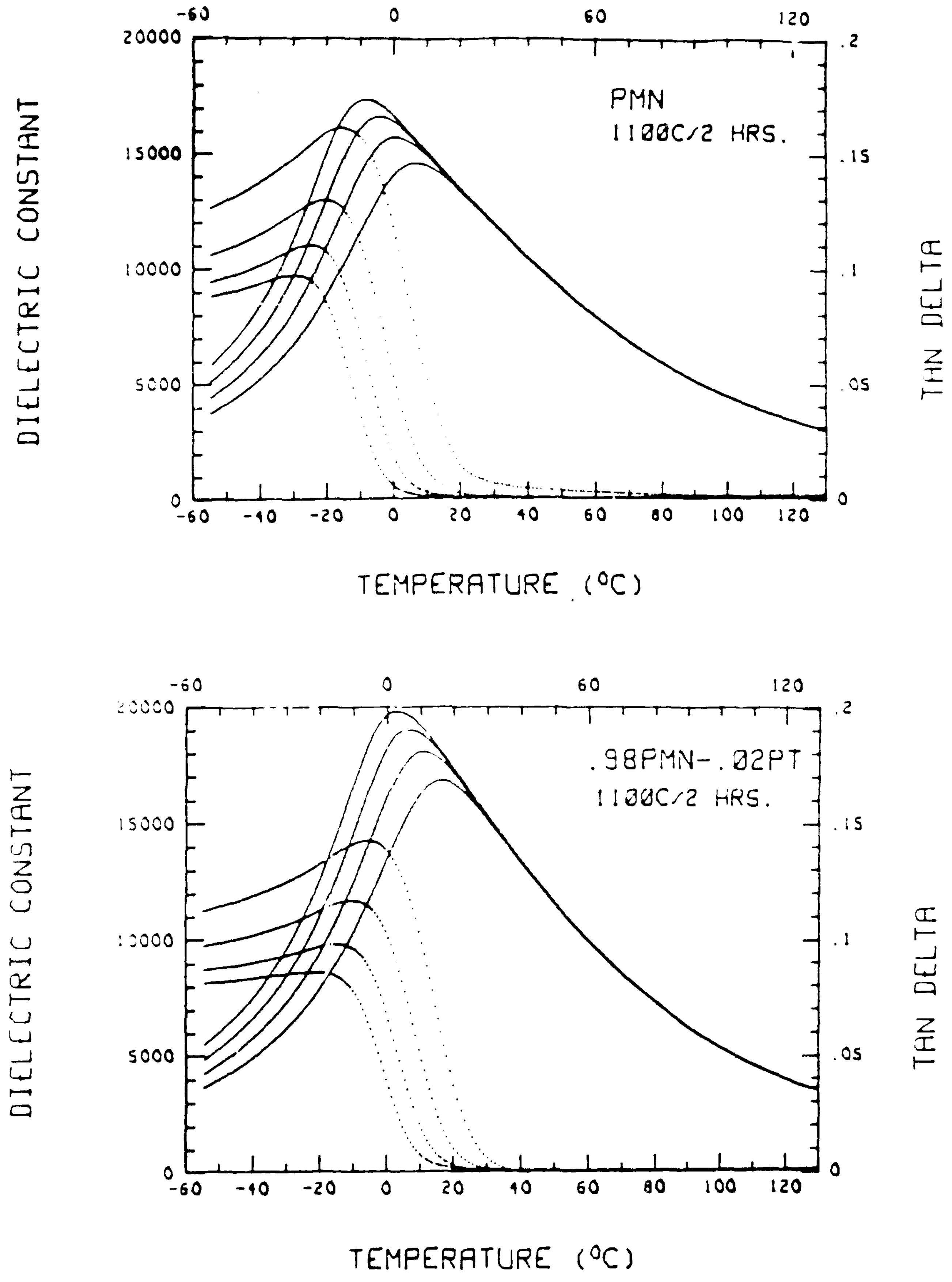
[6] Z.Y. Meng, U. Kumar, and L.E. Cross. "Electrostriction in Lead Lanthanum Zirconate-Titanate Ceramics," J. Amer. Ceram. Soc., 1042 (1988).

[7] J. von Cieminski and H. Beige. "High Signal Electrostriction in Ferroelectric Materials," J. Phys. D: Appl. Phys. 24, 1182-96, (1991).

List of Tables

Table I. Strain response for various electrostrictors at various temperatures and E-fields.
Note: Transverse strain (negative) (@ 0.1 Hz)

Composition	Temp.	Transverse Strain ($x10^{-4}$) 10 Kv/cm	20 Kv/cm	Hysteresis
PMN	28°C	0.4	1.4	Minimal
	12°C	0.6	1.6	"
	-6°C	0.8	2.1	"
	-14°C	0.9	2.2	"
	-34°C	1.2	2.6	"
PMN:PT (2% PT)	29°C	0.7	1.9	Minimal
	16°C	0.9	2.2	"
	13°C	0.9	2.3	"
	5°C	1.1	2.6	"
	-3°C	1.3	2.6	"
	-5°C	1.7	3.2	"
	-7°C	1.8	3.3	"
	-33°C	2.8	4.1	Significant
PMN:PT:La	15°C	0.6	1.4	Minimal
	8°C	0.6	1.6	"
	-1°C	0.7	1.8	"
	-3°C	0.7	1.9	"
	-6°C	0.7	1.9	"
	-8°C	0.8	1.9	"
	-12°C	1.3	2.8	Increased
	-21°C	1.4	3.0	"



Figures 1 and 2. Dielectric Behavior of PMN (Top) and .98 PMN-.02 PT (Bott 1 kHz, 10 kHz, and 100 kHz.

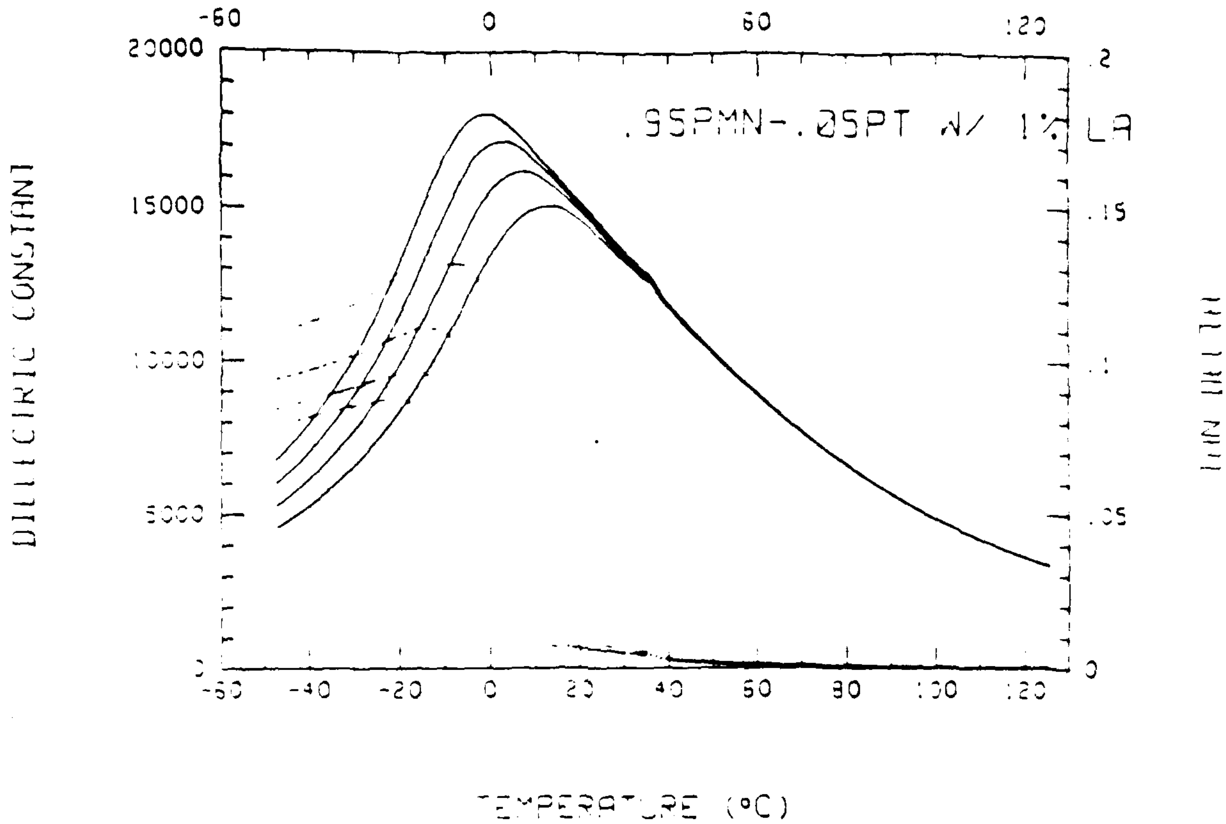


Fig. 3. Dielectric Behavior of .95 PMN-.05 PT:1% La at 100 Hz, 1 kHz, 10 kHz, and 100 kHz.

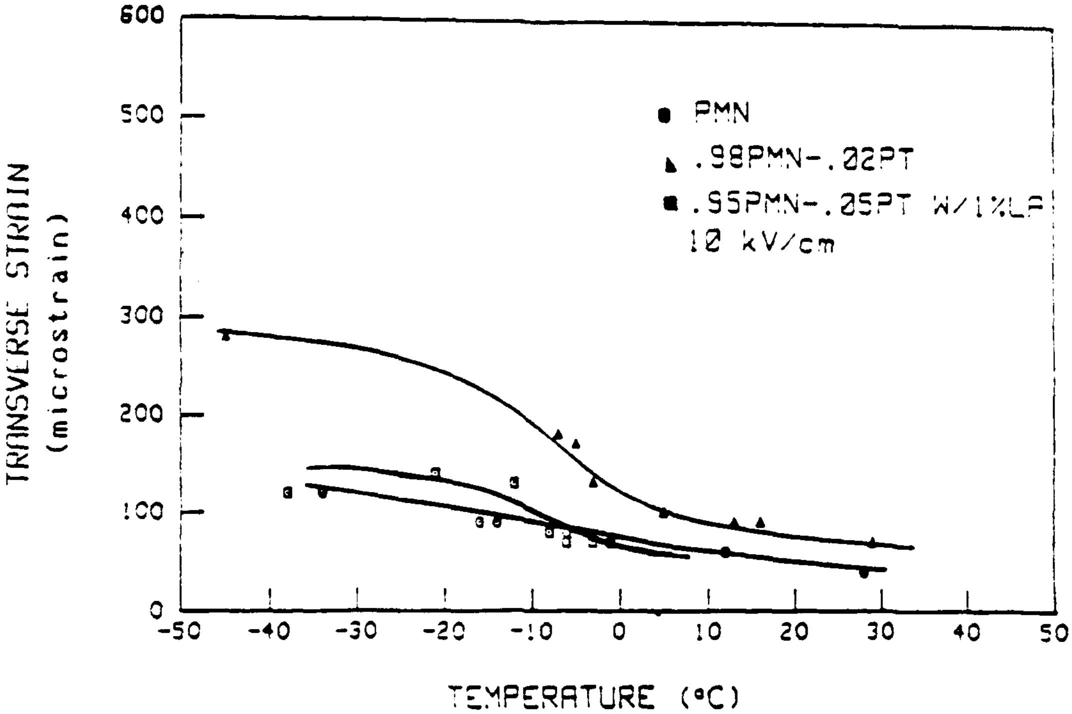
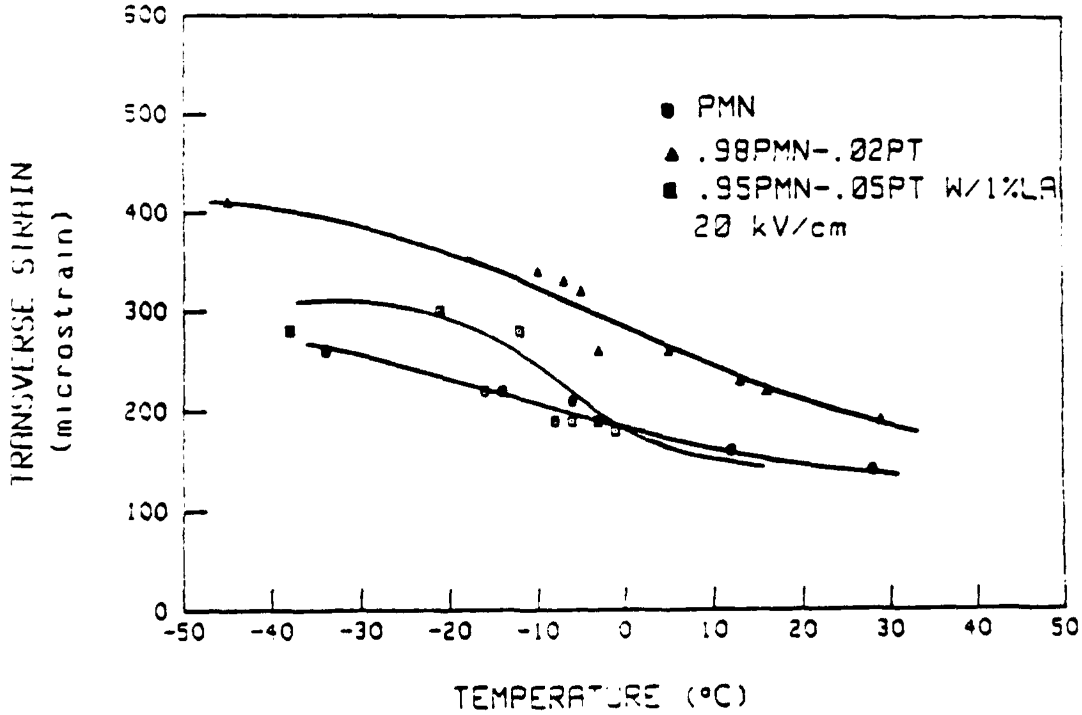


Fig. 4. Transverse strain (negative) as a function of temperature for various electrostrictors at 10 kV/cm and 20 kV/cm. [PMN, .98 PMN-.02 PT, .95 PMN-.05 PT:1% La].



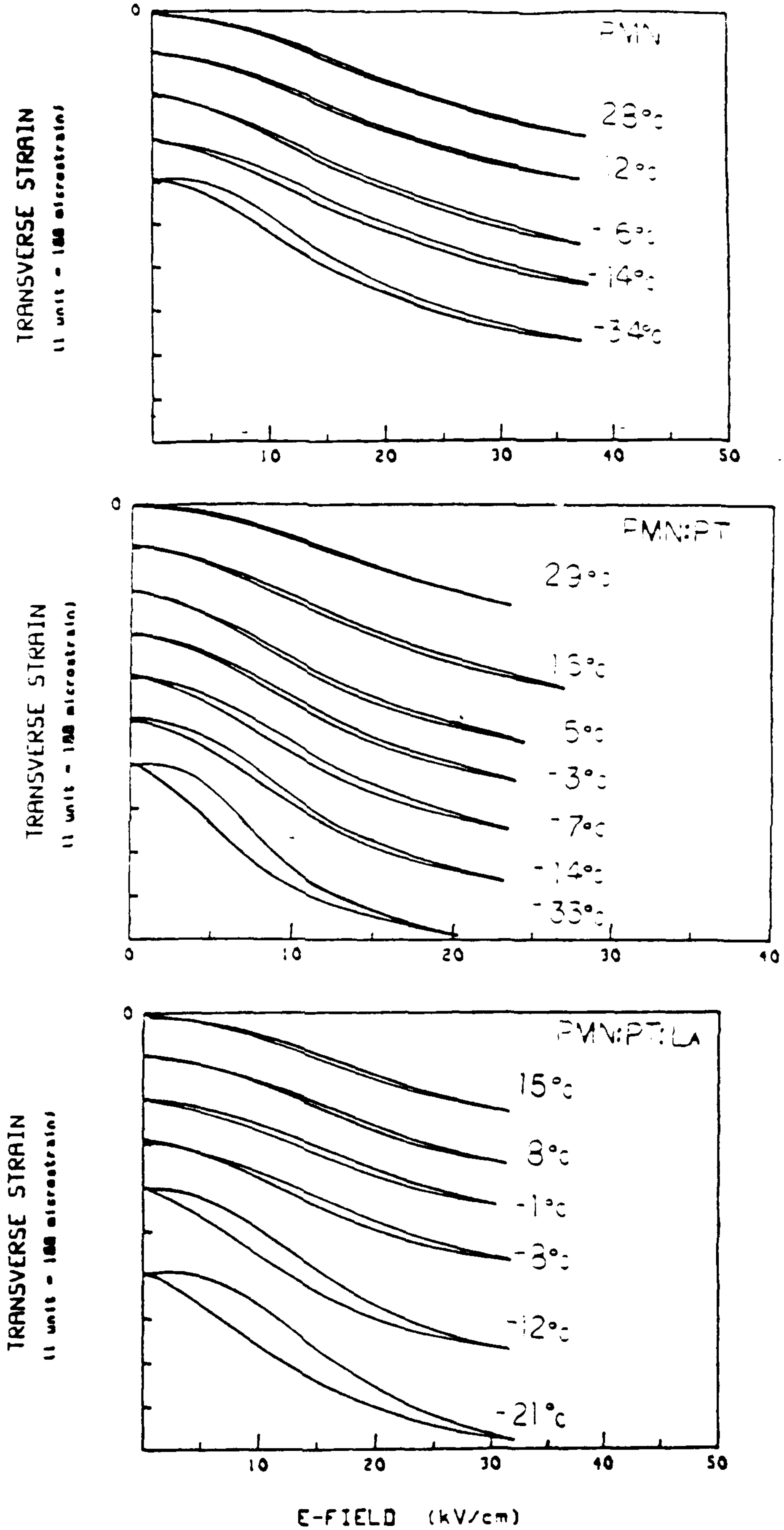


Fig. 5,6, and 7. Transverse strain (negative) as a function of electric field and temperature for PMN (Top), .98 PMN-.02 PT (middle), and .95 PMN-.05 PT:1% La (Bottom).

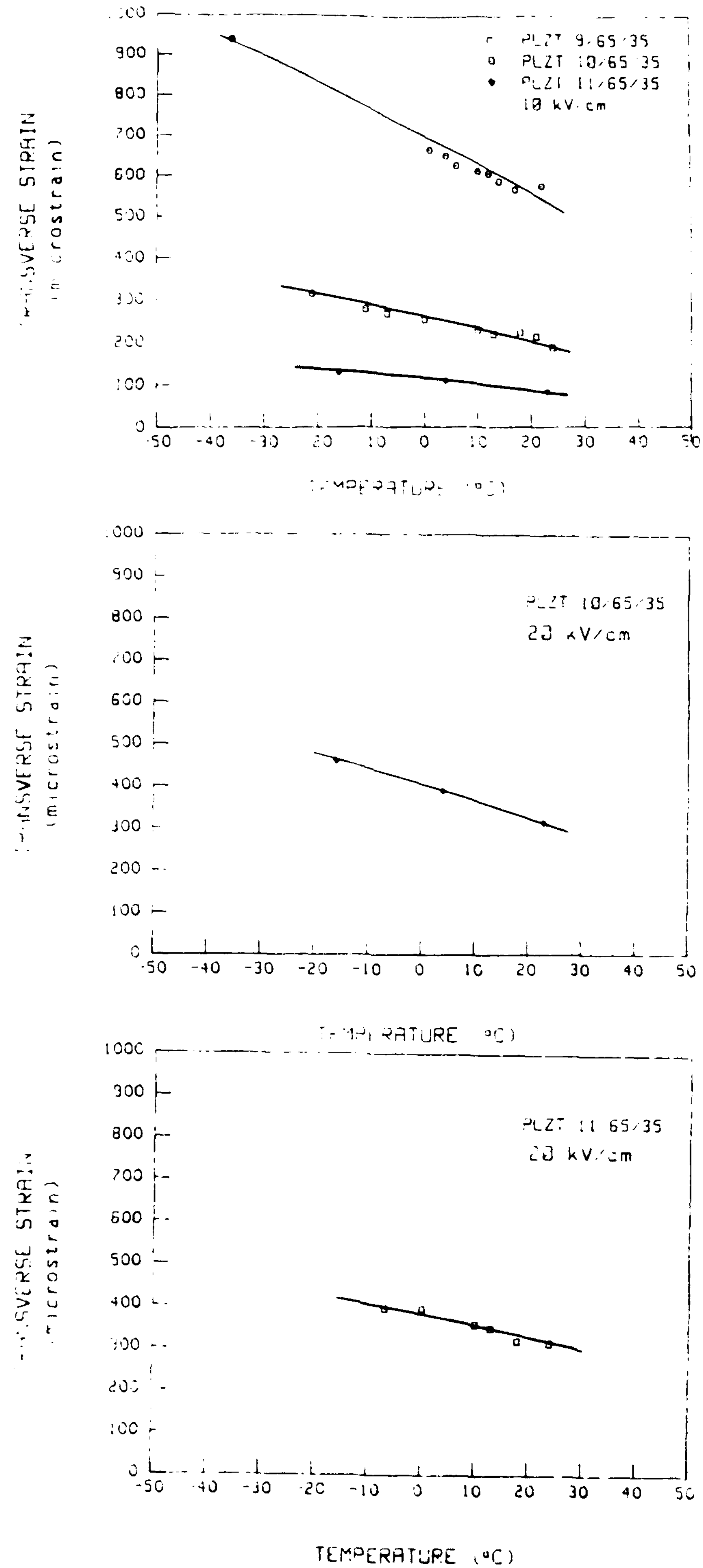
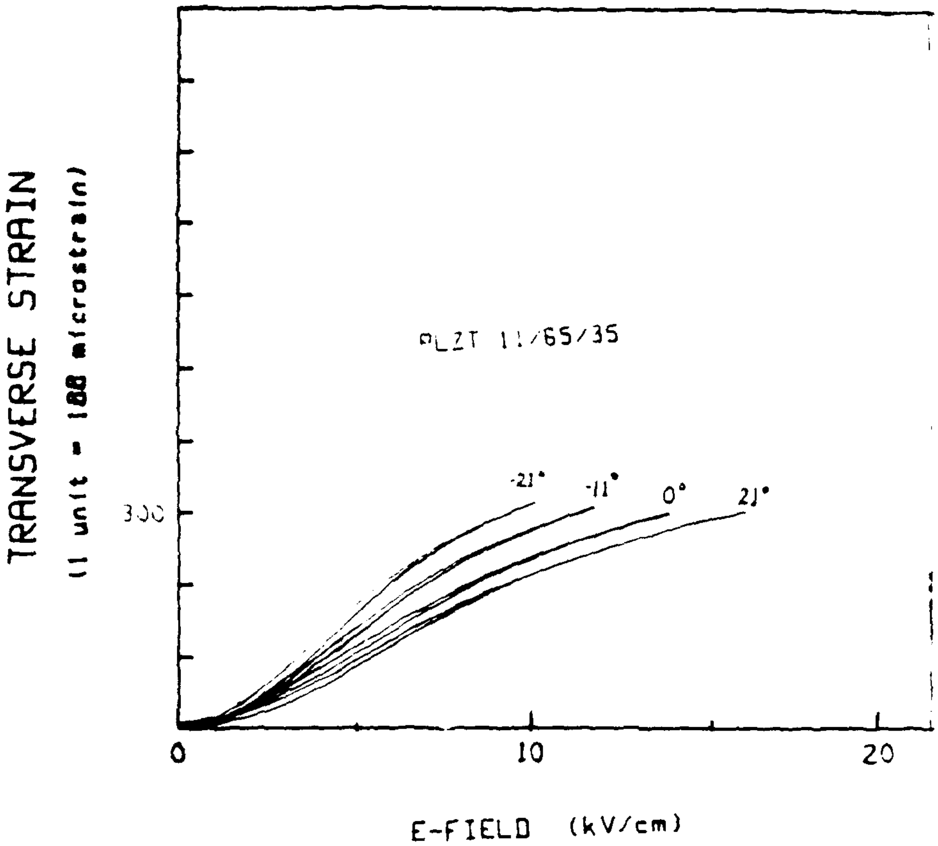


Fig. 8,9, and 10. **Transverse strain (negative) as a function of temperature for various PLZT electrostrictive compositions at 10 kV/cm (Top), for PLZT (10/65/35) at 20 kV/cm (middle), and for PLZT (11/65/35) at 20 kV/cm (Bottom).**



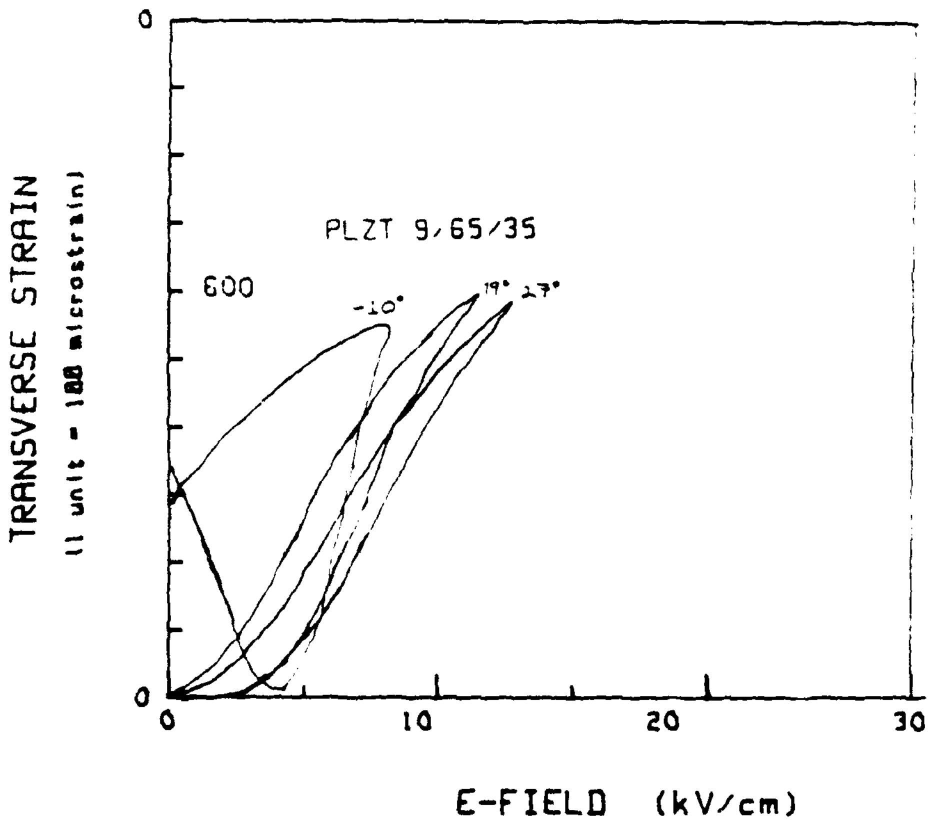


Fig. 11 and 12. (Top) Transverse strain (negative) with a constant maximum strain level (~300 μm) for PLZT (11/65/35) and bottom) with a constant maximum strain level (~600 μm) for PLZT (9/65/35) at various temperatures.

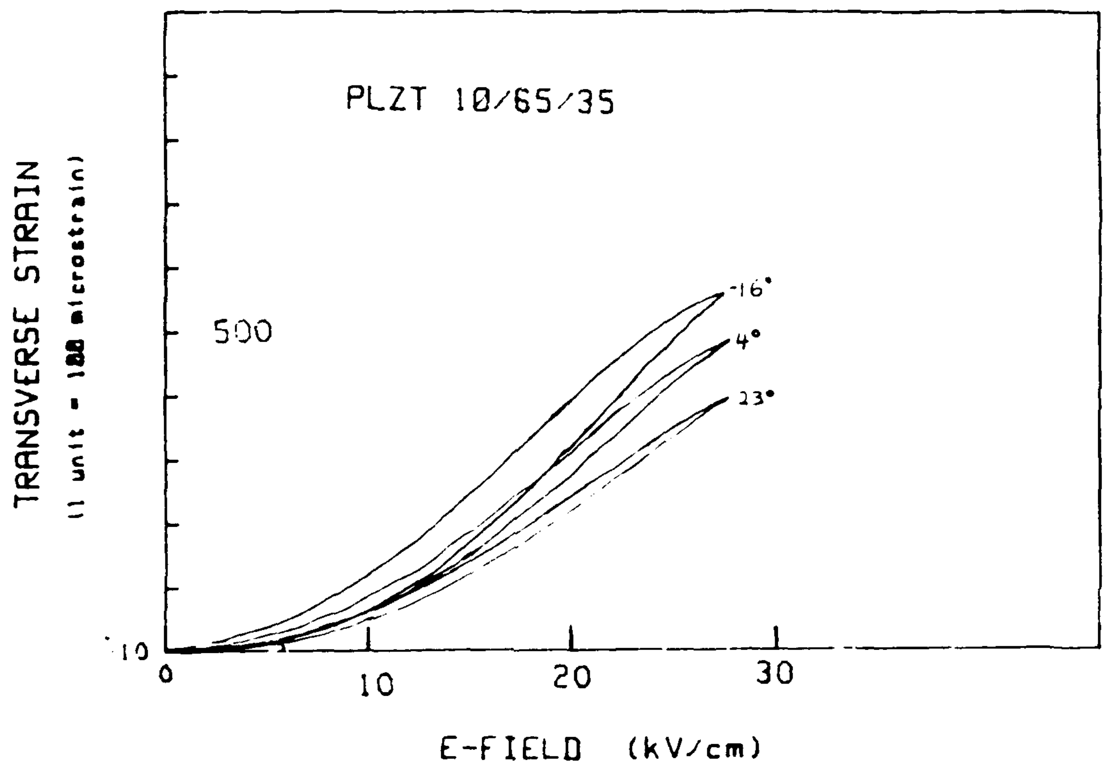


Fig. 13. Transverse strain negative with a constant maximum field (27.5 kV/cm) for PLZT (10/65/35).